

AMENDMENTS TO THE CLAIMS

Please substitute the following claims 116-118, 120-160, 162-166 and 184-195 as replacement claims for the previously-pending claims. In this Amendment C, claim 119 has been canceled, claims 116, 120-123, 130, 131, 135, 139, 140, 145-149, 151-154, 158-160, 162, 165 and 166 have been amended, and new claims 186-195 have been added.

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1-115. (canceled).

116. (amended) A method for providing materials to a parallel processing microsystem for identifying and characterizing materials that enhance a chemical reaction, the method comprising

simultaneously loading at least four materials into four or more microreactors such that the materials are individually resident in a reaction cavity of a separate microreactor, each of the at least four materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a volume of not more than about 3 ml for carrying out a chemical reaction, an inlet port in fluid communication with the reaction cavity for supplying one or more reactants to the reaction cavity, and an outlet port in fluid communication with the reaction cavity for discharging a reactor effluent from the reaction cavity,

each of the four or more reactors being formed in a plurality of laminae, wherein the at least four materials are loaded into the four or more microreactors as a material-containing laminate comprising a substrate and the at least four materials at separate portions of the substrate.

117. (original) The method of claim 116 wherein at least ten materials are simultaneously loaded into ten or more microreactors.

118. (original) The method of claim 116 wherein at least one-hundred materials are simultaneously loaded into one-hundred or more microreactors.

119. (canceled).

120. **(amended)** The method of claim 116 wherein ~~the four or more microreactors are formed in a plurality of laminae~~, the plurality of laminae in which the four or more microreactors are formed include including

the material-containing laminate as a material-containing first laminate comprising first and second surfaces in spaced, substantially parallel relationship to each other and the at least four materials,

a second laminate or composite substructure comprising a plurality of laminates, the second laminate or composite substructure comprising first and second surfaces in spaced, substantially parallel relationship with each other, and an array of four or more substantially coplanar wells arranged to correspond to the arrangement of the at least four materials of the first laminate, each of the wells having an interior surface defining an open-ended cavity at the first surface of the second laminate or composite substructure, the method further comprising

engaging the second surface of the ~~materials~~ material-containing first laminate and the first surface of the second laminate or composite substructure such that, taken together, the engaged laminae form an array of four or more microreactors defined by the interior surfaces of the wells and at least a portion of material-containing regions of the material-containing first laminate.

121. **(amended)** The method of claim 120 wherein the second surface of the ~~materials~~ material-containing first laminate and the first surface of second laminate are releasably engaged.

122. **(amended)** The method of claim 120 wherein the second surface of the ~~materials~~ material-containing first laminate and the first surface of second laminate are bonded.

123. **(amended)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

simultaneously loading at least four candidate catalyst materials into four or more microreactors of a chemical processing microsystem such that the at least four materials are individually resident in separate microreactors, thereby forming four or more material-containing microreactors, each of the at least four candidate catalyst materials comprising an inorganic

material, a metal-ligand or a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a volume of not more than 3 ml,

simultaneously supplying one or more reactants to each of the four or more microreactors,

simultaneously contacting each of the at least four candidate catalyst materials with the one or more reactants in the four or more microreactors under reaction conditions for the reaction of interest,

simultaneously discharging a reactor effluent from each of the four or more material-containing microreactors,

evaluating the at least four candidate catalyst materials for catalytic activity for the chemical reaction of interest.

124. **(original)** The method of claim 123 wherein the amount of each of the at least four materials loaded into the microreactors is not more than about 5 mg.

B/ 125. **(original)** The method of claim 123 wherein the amount of each of the at least four materials loaded into the microreactors is not more than about 1 mg.

126. **(original)** The method of claim 123 wherein at least ten materials are simultaneously loaded into ten or more microreactors.

127. **(original)** The method of claim 123 wherein at least one-hundred materials are simultaneously loaded into one-hundred or more microreactors.

128. **(original)** The method of claim 123 wherein the at least four materials are simultaneously loaded into the four or more microreactors as an array of candidate materials, the array comprising a substantially planar substrate and four or more materials at separate portions of the substrate.

129. **(original)**. The method of claim 123 further comprising simultaneously unloading the reactant-contacted materials from the microreactors in which they reside after the chemical reaction of interest.

130. **(amended)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

loading at least four candidate catalyst materials into four or more microreactors formed in a plurality of laminae such that the at least four materials are individually resident in separate microreactors, each of the at least four candidate catalyst materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a volume of not more than 3 ml,

simultaneously supplying one or more reactants to each of the four or more microreactors,

simultaneously contacting each of the at least four candidate catalyst materials with the one or more reactants in the four or more microreactors under reaction conditions for the reaction of interest,

simultaneously discharging a reactor effluent from each of the four or more material-containing microreactors, and

evaluating the at least four candidate catalyst materials for catalytic activity for the chemical reaction of interest,

the at least four candidate catalyst materials being loaded into the four or more microreactors without affecting the structural integrity of a fluid distribution system through which the one or more reactants are supplied to the microreactors or through which one or more reactor effluents are discharged from the microreactors.

131. **(amended)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

loading at least four candidate catalyst materials into four or more microreactors at a time  $t_1$  such that the at least four materials are individually resident in separate microreactors, each of the at least four candidate catalyst materials comprising an inorganic material, a metal-ligand or

a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a volume of not more than 3 ml,

simultaneously supplying one or more reactants to each of the four or more microreactors,

simultaneously contacting each of the at least four candidate catalyst materials with the one or more reactants in the four or more microreactors under reaction conditions for the reaction of interest,

simultaneously discharging a reactor effluent from each of the four or more material-containing microreactors, and

evaluating the at least four candidate catalyst materials for catalytic activity for the chemical reaction of interest at a time  $t_2$ , the difference in time,  $t_1-t_2$ , being less than about 3 hours.

132. **(original)** The method of claim 131 wherein the difference in time,  $t_1-t_2$ , is not more than about 1 hr.

133. **(original)** The method of claim 131 wherein the difference in time,  $t_1-t_2$ , is not more than about 30 minutes.

134. **(original)** The method of claim 131 wherein the difference in time,  $t_1-t_2$ , is not more than about 15 minutes.

135. **(amended)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

loading at least ten candidate catalyst materials into ten or more microreactors such that the at least ten materials are individually resident in separate microreactors, each of the at least ten candidate catalyst materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the ten or more microreactors comprising a surface defining a reaction cavity having a volume of not more than 3 ml,

simultaneously supplying one or more reactants to each of the ten or more microreactors,

simultaneously contacting each of the at least ten candidate catalyst materials with the one or more reactants in the ten or more microreactors under reaction conditions for the reaction of interest,

simultaneously discharging a reactor effluent from each of the ten or more material-containing microreactors, and

evaluating the at least ten candidate catalyst materials for catalytic activity for the chemical reaction of interest, the at least ten materials being evaluated for catalytic activity at a throughput of not less than about 10 materials / hour.

136. **(original)** The method of claim 135 wherein the at least ten materials are evaluated for catalytic activity at a throughput of not less than about 100 materials / hour.

137. **(original)** The method of claim 135 wherein the at least ten materials are evaluated for catalytic activity at a throughput of not less than about 1000 materials / hour.

3 138. **(original)** The method of claim 135 wherein the at least ten materials are at least ten different materials.

139. **(amended)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

loading at least four candidate catalyst materials into four or more microreactors such that the at least four materials are individually resident in separate microreactors, each of the at least four candidate catalyst materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a geometry and having a volume of not more than 3 ml,

simultaneously supplying one or more reactants to each of the four or more microreactors,

simultaneously contacting each of the at least four candidate catalyst materials with the one or more reactants in the four or more microreactors,

selecting the geometry of the reaction cavity and controlling the reaction conditions in the four or more microreactors such that the reactant residence time,  $\tau_{res}$ , is longer than the diffusion period,  $\tau_{diff}$ , for the reaction cavity,

simultaneously discharging a reactor effluent from each of the four or more material-containing microreactors, and

evaluating the at least candidate catalyst four materials for catalytic activity for the chemical reaction of interest.

140. **(amended)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

loading at least four candidate catalyst materials into four or more microreactors of a chemical processing microsystem such that the at least four materials are individually resident in separate microreactors, each of the at least four candidate catalyst materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a volume of not more than 10  $\mu\text{l}$ ,

simultaneously supplying one or more reactants to each of the four or more microreactors,

simultaneously contacting each of the at least four candidate catalyst materials with the one or more reactants in the four or more microreactors,

simultaneously discharging a reactor effluent from each of the four or more material-containing microreactors into separate four or more microseparators of the chemical processing microsystem,

simultaneously separating one or more components of the reactor effluents in the four or more microseparators, and

evaluating the at least four candidate catalyst materials for catalytic activity for the chemical reaction of interest.

141. **(original)** The method of claims 116, 123, 130, 131, 139 or 140 wherein the at least four materials are at least four different materials.

142. **(original)** The method of claims 116, 123, 130, 131, 139 or 140 wherein the reaction cavity of each of the four or more material-containing microreactors has a volume of not more than about 100  $\mu$ l.

143. **(original)** The method of claims 116, 123, 130, 131, 139 or 140 wherein the reaction cavity of each of the four or more material-containing microreactors has a volume of not more than about 10  $\mu$ l.

144. **(original)** The method of claims 116, 123, 130, 131, 139 or 140 wherein the reaction cavity of each of the four or more material-containing microreactors has a volume of not more than about 1  $\mu$ l.

145. **(amended)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

loading at least twenty five ~~four~~ candidate catalyst materials into twenty five ~~two-hundred-fifty~~ or more microreactors using an automated material handling system such that the at least twenty five ~~four~~ materials are individually resident in separate microreactors, each of the at least twenty five ~~four~~ candidate catalyst materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the twenty five ~~two-hundred-fifty~~ or more microreactors comprising a surface defining a reaction cavity having a volume of not more than 3 ml,

simultaneously supplying one or more reactants to each of the twenty five ~~two-hundred-fifty~~ or more microreactors,

simultaneously contacting each of the at least twenty five ~~four~~ candidate catalyst materials with the one or more reactants in the twenty five ~~two-hundred-fifty~~ or more microreactors under reaction conditions for the reaction of interest,

simultaneously discharging a reactor effluent from each of the twenty five ~~two-hundred-fifty~~ or more material-containing microreactors, and

evaluating the at least twenty five ~~four~~ candidate catalyst materials for catalytic activity for the chemical reaction of interest.



146. **(amended)** The method of claim 145 further comprising unloading the at least twenty five ~~four~~ reactant-contacted materials from the microreactors in which they reside, and

loading a second set of at least twenty five ~~four~~ materials into the twenty five ~~four~~ or more microreactors of the chemical processing microsystem using the automated material handling system such that the second set of at least twenty five ~~four~~ materials are individually resident in separate microreactors.

147. **(amended)** The method of claim 146 wherein the at least twenty five ~~four~~ candidate materials are loaded simultaneously into the twenty five ~~four~~ or more microreactors, and the reactant-contacted candidate materials are unloaded simultaneously therefrom.

148. **(amended)** The method of claim 146 wherein the at least twenty five ~~four~~ materials are loaded sequentially into the twenty five ~~four~~ or more microreactors, and the reactant-contacted candidate materials are unloaded therefrom.

149. **(amended)** The method of claim 146 wherein ~~one or more of the step steps of loading the at least four candidate materials into the four or more microreactors, unloading the reactant-contacted candidate materials from the microreactors, and loading a second set of at least four candidate materials into the four or more microreactors~~ are is effected using the automated material handling system.

150. **(original)** The method of claim 145 wherein the one or more reactants are gaseous reactants.

151. **(amended)** The method of claim 145 wherein the at least twenty five ~~four~~ materials are contacted with the one or more reactants under a set of reaction conditions, the method further comprising controlling the reaction conditions to be substantially the same in each of the twenty five ~~two hundred fifty~~ or more microreactors.

152. **(amended)** The method of claim 145 wherein the at least twenty five ~~four~~ candidate materials are contacted with the one or more reactants under a set of reaction conditions; the method further comprising controlling the reaction conditions to be substantially the same in at least a subset ~~each~~ of the twenty five ~~two hundred fifty~~ or more microreactors according to one or more control protocols selected from the group consisting of:

controlling the temperature to be not less than about 100 °C and to be substantially the same in at least four of the twenty-five ~~two hundred fifty~~ or more microreactors,

controlling the pressure to range from about 1 atm to about 200 bar and to be substantially the same in at least four of the twenty five ~~two hundred fifty~~ or more microreactors,

controlling the residence time to range from about 1  $\mu$ sec to about 1 hour and to be substantially the same in at least four of the twenty five ~~two hundred fifty~~ or more microreactors, and

controlling the reactant flow rate to be substantially the same through at least four of the twenty five ~~two hundred fifty~~ or more microreactors.

153. **(amended)** The method of claim 145 wherein the at least twenty five ~~four~~ materials are contacted with the one or more reactants under a set of reaction conditions, the method further comprising controlling the reaction conditions to be varied between at least two of the twenty five ~~two hundred fifty~~ or more microreactors.

154. **(amended)** The method of claim 145 wherein the at least twenty five ~~four~~ materials are contacted with the one or more reactants under a set of reaction conditions, the method further comprising controlling the reaction conditions to be varied between at least two of the twenty five ~~two hundred fifty~~ or more microreactors according to one or more control protocols selected from the group consisting of:

controlling the temperature to be varied between at least two of the twenty five ~~two hundred fifty~~ or more microreactors,

controlling the pressure to be varied between at least two of the twenty five ~~two hundred fifty~~ or more microreactors,

controlling the residence time to be varied between at least two of the twenty five ~~two hundred fifty~~ or more microreactors, and

controlling the reactant flow rate to be varied through at least two of the twenty five ~~two-hundred-fifty~~ or more microreactors.

155. **(original)** The method of claim 145 wherein the candidate materials are a film of material formed on a surface of the reaction cavity.

156. **(original)** The method of claim 145 wherein the microsystem comprises four-hundred or more microreactors.

157. **(original)** The method of claim 145 wherein the microsystem comprises one-thousand or more microreactors.

158. **(amended)** The method of claim 145 wherein different candidate materials are individually resident in the separate reaction cavities of at least 90% of the twenty five ~~two-hundred-fifty~~ or more microreactors.

159. **(amended)** The method of claim 145 wherein at least twenty five ~~four~~ materials are evaluated for catalytic activity according to one or more analytical protocols selected from the group consisting of

determining catalytic activity by analytical measurement of the reactor effluent,  
determining catalytic activity by *in situ* analytical measurement,  
determining catalytic activity by serial analytical measurement,  
determining catalytic activity by parallel analytical measurement, and  
determining catalytic activity of a subset of the at least four materials by parallel analytical measurement.

160. **(amended)** The method of claim 145 wherein at least twenty five ~~four~~ materials are evaluated for catalytic activity according to one or more analytical protocols selected from the group consisting of

determining catalytic activity by parallel or serial gas chromatography of the reactor effluents,

determining catalytic activity by separating one or more components of the reactor effluents and determining the presence, absence or amount of the separated one or more components,

determining catalytic activity by adsorbing one or more components of at least four reactor effluents onto an adsorbent material, and determining the presence, absence or amount of adsorbed component,

determining catalytic activity by adsorbing one or more components of at least four reactor effluents onto an adsorbent material, desorbing an adsorbed component, and determining the presence, absence or amount of desorbed component, and

determining catalytic activity by determining the amount of a reaction product formed by the chemical reaction of interest.

161. (canceled).

3 162. (amended) A method for evaluating or optimizing process conditions for a chemical reaction of interest, the method comprising

simultaneously supplying one or more reactants to each of four or more microreactors, each of the microreactors comprising a surface defining a reaction cavity having a volume of not more than about 10  $\mu$ l for carrying out a chemical reaction, an inlet port in fluid communication with the reaction cavity, and an outlet port in fluid communication with the reaction cavity,

controlling a first set of reaction condition ~~conditions~~ to be substantially identical in each of the microreactors,

controlling a second set of reaction condition ~~conditions~~ to be varied between two or more of the microreactors,

simultaneously discharging a reactor effluent from each of the four or more microreactors to four or more microseparators, each of the microseparators comprising a surface defining a separation cavity for separating at least one component of a reactor effluent, an inlet port in fluid communication with the outlet port of one of the microreactors for receiving the reactor effluent therefrom, and an outlet port in fluid communication with the separation cavity, and

simultaneously discharging the separated effluent from each of the microseparators, and evaluating the effect of varying the second set of reaction conditions.

163. **(original)** The method of claim 162 wherein the four or more microreactors are formed in a plurality of laminae and the four or more microseparators are formed in a plurality of laminae.

164. **(original)** The method of claim 162 wherein the reaction cavity of each of the at least four candidate material-containing microreactors has a volume of not more than about 1  $\mu$ l.

165. **(amended)** The method of claim 162 wherein the first and second ~~set of~~ reaction conditions are independently selected from the group consisting of temperature, pressure, residence time and flow rate.

166. **(amended)** The method of claim 162 wherein the first and second ~~set of~~ reaction conditions are independently selected from the group consisting of temperature, pressure, and residence time.

167-183. (canceled).

184. **(original)** A method for effecting a microscale chemical reaction, the method comprising

supplying one or more reactants for a chemical reaction of interest to a microreactor, the microreactor comprising a surface defining a reaction cavity having a volume of not more than about 10  $\mu$ l for carrying out a chemical reaction, an inlet port in fluid communication with the reaction cavity for supplying one or more reactants thereto, and an outlet port in fluid communication with the reaction cavity for discharging one or more reaction products therefrom, and

converting the one or more reactants to one or more reaction products in the reaction cavity,

the reactants residing in the reaction cavity under process conditions effective for the chemical reaction of interest for a residence time,  $\tau_{\text{res}}$ , that is longer than the diffusion period,  $\tau_{\text{diff}}$ , for the reaction cavity under such process conditions.

185. **(original)** The method of claim 184 wherein the microreactor further comprises a catalyst within the reaction cavity for catalyzing the chemical reaction of interest.

186. **(new)** The method of claim 123 wherein the one or more reactants are simultaneously supplied to each of the four or more microreactors through a fluid distribution system, the fluid distribution system providing fluid communication from one or more reactant sources to the reaction cavities of each of the four or more microreactors through a microfluidic fluid-supply manifold.

187. **(new)** The method of claims 123 or 186 wherein the reactor effluent is simultaneously discharged from each of the four or more material-containing microreactors through an effluent distribution system providing fluid communication from the reaction cavity of each of the four or more microreactors to one or more effluent sinks through a microfluidic effluent-distribution manifold.

188. **(new)** The method of claim 186 wherein the fluid distribution system is effective for supplying one or more gaseous reactants through the microfluidic fluid-supply manifold.

189. **(new)** The method of claim 123 further comprising controlling the temperature of the reaction cavities of each of the four or more microreactors to be above 100 °C during the chemical reaction of interest.

190. **(new)** The method of claim 123 further comprising controlling the temperature of the reaction cavities of each of the four or more microreactors to be above 200 °C during the chemical reaction of interest.

191. **(new)** The method of claim 123 further comprising controlling the temperature of the reaction cavities of each of the four or more microreactors to range from about 100 °C to about 500 °C during the chemical reaction of interest.

192. **(new)** The method of claim 123 further comprising controlling the temperature of the reaction cavities of each of the four or more microreactors to range from about 100 °C to about 800 °C during the chemical reaction of interest.

193. **(new)** The method of claim 145 wherein the automated material handling system is adapted for simultaneously loading candidate catalyst materials into the microreactors.

194. **(new)** The method of claim 145 wherein the automated material handling system is adapted for serially loading candidate catalyst materials into the microreactors.

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195. **(new)** The method of claim 145 wherein the automated material handling system is adapted for automatically or semiautomatically loading candidate catalyst materials into the microreactors.

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